Analysis of ETC or Classical Manometric closed vessel tests with coupling of thermodynamic equilibrium calculations: combustion rate, Energy losses

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ANALYSIS OF ETC OR CLASSICAL MANOMETRIC CLOSED VESSEL TESTS WITH COUPLING OF THERMODYNAMIC EQUILIBRIUM CALCULATIONS: COMBUSTION RATE, ENERGY LOSSES

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We performed closed vessel tests to study the combustion of several gun propellants ignited classically or by ETC. We analysed the results to estimate the combustion rates and energy losses to the bomb wall. The classical STANAG 4115 method for the determination of the combustion rate of solid gun propellants classically ignited was adapted for ETC conditions. The experimental combustion rates obtained were compared with existing more fundamental expressions of this rate by coupling thermodynamic equilibrium calculations with the experimental results. This coupling allows also the estimation of energy losses to the bomb wall. As this research is still underway, we present the more significant results on a double base propellant (NC/NGL) and their present interpretation and limits.

INTRODUCTION

As part of the French ETC Programme, we performed and analysed closed vessel tests to estimate combustion rates of several gun propellants ignited classically or by ETC and the energetic interactions involved for modelling purposes. The objective of the modelling is to establish easy-to-use more fundamental expressions validated with experimentation, where possible. We present the current state of our analysis and interpretation on the more significant results we have on a NC/NGL propellant.

The experimental combustion rates obtained by this method were then compared with existing more fundamental expressions some of which require the knowledge of the nature of the gas-phase (composition, pressure and temperature). For this purpose, the gas phase is considered to follow a succession of thermodynamic equilibria which are calculated using the code ALJAN-EP [1]. These calculations enable estimation of the transient nature of the gas phase. Expressions for combustion rate can then be formulated and energy losses to the bomb wall estimated.

EXPERIMENTAL AND ANALYSIS

The combustion experiments were performed in a 460 cm^3 closed vessel, including a plasma torch 8 cm long and 1 cm in diameter, to generate plasma corresponding to an injected energy of up to 40 kJ. The classical ignition is the same configuration as the above employing the hot wire method and a small amount of pyrotechnic igniting material.

Determination of experimental combustion rates
Principles

The classical STANAG 4115 method and its related assumptions for the determination of the combustion rate of solid gun propellant, from manometric closed vessel firings, classically ignited, was simply adapted for ETC conditions. For ETC ignition, we assume now that the initial pressure – when the mass fraction of burnt propellant is still null – is that pressure obtained during the plasma injection phase, where the pressure versus time curve exhibits a slope change. This point is supposed to correspond to the beginning of the propellant combustion phase.

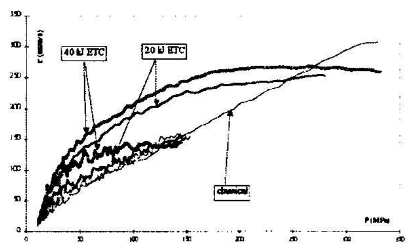

Fig. 1: STANAG type combustion rate of double base propellant at LD 0.15 and LD 0.3.

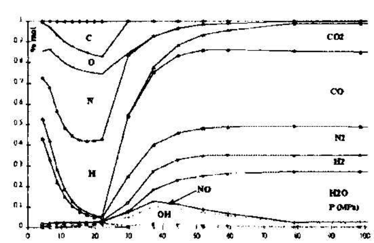

Fig. 2: 40 kJ ETC ignition: Cumulated species molar fraction of the gas phase vs. pressure.

Figure 1, shows the combustion rate of a double base propellant at loading densities of 150 (LD 0.15) and 300 kg/m^3 (LD 0.30) ignited with 20 kJ (bold line) and 40 kJ (o) ETC ignition, and by classical ignition (thin line). The three curves stopping at 150 MPa apply to a LD 0.15 and the 300 MPa readings are for LD 0.3. For both LD, the lower curve is for classic ignition, the middle curve is for 20 kJ ETC and the upper curve for 40 kJ. Applying the same numerical smoothing, the combustion rates present less oscillations at high LD than at the lower level. 20 kJ ETC and classic ignition seem very similar for LD 0.15, so in these conditions they could be considered as equivalent. At higher LD, there is an unusual looking combustion rate up to 250 MPa. So, in the following text, we will consider only 40 kJ ETC and classical ignition at LD 0.3.

Transient composition of the gas phase

The hypothesis that the gas phase follows a succession of thermodynamic equilibria is generally considered as correct for classical ignition above 4000 psi (28 MPa). We also performed some elementary kinetic calculations on N2-O2 mixes which allow us to consider that this hypothesis could be correct above 10 MPa for CHON products ignited by ETC.

Thermodynamic equilibrium calculation allows the estimation of the transient composition of the gas phase. Assuming that there is no combustion of the GP until the end of the plasma injection, as suggested by the results of previous lower energy interrupted tests, we carried out numerical simulation of the cumulated molar fraction of the gas phase versus pressure, presented in Figure 2, for 40 kJ ETC ignition, for LD 0.3 up to 100 MPa. Up to 20 MPa, the gas phase in the vessel itself (generated by the plasma injected from the torch and the whole of the initial air) is essentially atomic – with very few ions. As the propellant starts to burn, it cools down this gas phase, then the molecular formation becomes possible and stabilises itself toward the composition of the classically ignited propellant. Note that this assumption of no reaction of the GP until the end of plasma injection for this numerical simulation does not correspond with the experimental results. For the latter, GP seems to start to react at around 10 MPa when 20 kJ has already been injected. Our current estimates are that 5% of the propellant is burnt at the end of the 40 kJ plasma injection. Further results are required to clarify this point.

Estimation of energy losses and flux to the bomb wall
Principle

The internal energy of the gas phase in the bomb is determined at the experimental pressure according to the assumptions implied by STANAG 4115. In the absence of leakage of matter, its derivative with time maybe considered as due to a global energetic flux Φ_E to the bomb wall of surface S.

The global energy losses to the wall Q are the difference between the internal energy of the gas phase in the vessel itself, which is composed of the fraction of burnt propellant, a small amount of polyethylene (PE: plasma torch wall), and the initial air and their internal energies of formation $Q = U - \Delta U^0{}_f(\text{GP, air, PE})$.

Application and discussion

While awaiting further results to clarify the difficult questions regarding ETC ignition mentioned above, we present in this work an attempted application to classical ignition to get an estimation of the energy losses and corresponding flux for classical ignition presented in Figure 3. The energy losses (squares) seem reasonable as first analysis. The energetic flux (triangles) is proportional to its time derivative. The reduction of the energetic flux after the inflexion of the pressure is puzzling, as the experimental temperature (or adiabatic temperature) during the pressure rise is roughly constant at around 3200 K ± 6%

(3470 K ± 5%). The latter could perhaps be explained for instance by (1) an increase of energetic transfer while the grain is burning, or (2) the presence of non-equilibrium particles, as detected by spectroscopic measurements, taken by SNPE [2], in the reaction zone of burning propellants, or (3) and much more likely as simply a numerical artefact related to one of the STANAG assumptions used as the tool of this analysis (i.e. the relationship between experimental force, burnt mass of the propellant and the experimental pressure).

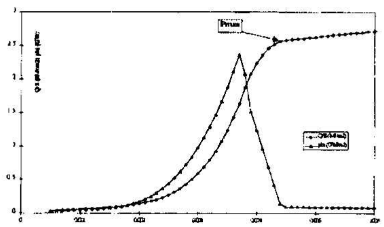

Fig. 3: Energy losses and related flux for classical ignition.

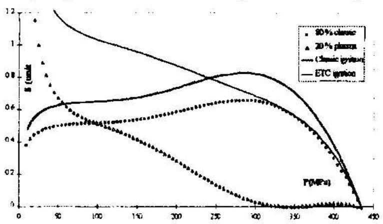

Fig. 4: Possible decomposition of grain burning surfaces of ETC ignition vs. pressure.

Application of existing models to the combustion rate

Several more or less fundamental expressions or results have been recently published on the combustion rate of gun propellants which may be categorised as below, according to physical or chemical effects and their related physical or chemical models which impose their rate upon the global combustion model:

- ablative; by energetic transfer to the surface of the propellant (with faster kinetics of post reactions of the surface – pyrolisis, state change or mechanical effect – or in the gas phase – not considered in this work);
- essentially radiative which needs to be coupled with other physical models;
- with slowest kinetic burning reaction of the surface layer of the propellant;
- different behaviours of combustion of the gun propellant grains when ignited by plasma compared to classical ignition: plasma modified behaviour for part of the grains, classical for the rest.

Moreover, in addition to classical pressure laws, several other semi-empirical models exist including those for the temperature of the gas phase or the electrical power injected into the plasma.

1 – Ablative models

We developed several ablative model whose generic expression is $r_{ab} = \frac{1}{\rho_m} \cdot \frac{\Phi_{th}}{E_{ab}}$ with a density ρ_m of the gun propellant at its initial temperature, an energetic flux Φ_{th} and a propellant ablation energy of E_{ab}. In the recent ablative models published by Lowe [4–5], Eisenreich et al. [6] and the one we presented at 18 ISB for classical combustion rate [3], the expressions of combustion rate are very similar.

Discussion on ablation energy and energetic flux

Currently as research is still ongoing, we lack elements in order to discriminate the partitioning of the energetic flux, which can be, as assumed in the literature, radiative (for the fraction transformed into thermal flux to the surface), conductive, convective or even mechanical!

For ablation energy E_{ab}, some of these models include a latent heat of state change or pyrolysis, but reduce or may reduce their expression to a simple heating of the propellant surface to a temperature T_s as $E_{ab} = c(T_s-T_o)$ with T_o the initial temperature of the gun propellant. The latter expression allows the estimation of E_{ab} from the results of classical ignition shots at several initial temperatures, as we have previously shown for a LOVA propellant [3], or as Eisenreich et al. [7] presented for JA2 – more similar than the LOVA to the double base propellant of this research.

Potential applications

As we do not yet have the necessary results, we can investigate a solution path considering as a first approximation that JA2 is similar to our propellant, and analyse the data published in [7]. We find for JA2, an ablation energy at a supposed initial temperature To = 293 K of 1.5 MJ/kg which corresponds, with a c estimated at 3.6 kJ/kg/K, to a heating of 417 K of the surface of the propellant up to the temperature of Ts = 710 K, coherent with ICT work. Further to the initial approximation, we can suppose that this ablation energy has no reason to change if the slowest process in the combustion is the heating to ablation temperature of the surface of the propellant. Once at this temperature, the propellant may melt, or be vaporised or pyrolysed, with a latent or reaction heat. This reaction, analogous to the type of reaction proposed by SNPE [2], could even be athermal, corresponding to our solid propellant at Ts = 710 K, giving first decomposition products as gaseous CH_2O and N2 and solid graphite at T = 1042 K.

If the ablation energy is constant, the ablation energetic flux $\Phi_{th} = \rho_m.E_{ab}.r_{ab}$ is simply proportional to the combustion rate and the correlation of this flux with the nature of the gas phase may then be investigated.

Current application to classical combustion

We tested our ablative model for the gun propellant combustion rate r, depending for a given propellant on constants K and n, on pressure P and on collision rate Z of particles of the gas phase, per unit of time and surface, i.e. $r(P,T,n_X) = KP^nZ$. We looked for two possible origins of the energetic source allowing the ablative feedback to the surface of the GP: (1) in the equilibrium gas phase and (2) in the possible hottest region of the vapour layer supposed to correspond to adiabatic equilibrium. These results shows that it is feasible to easily obtain such expressions but do not enable yet identification of the best physical model.

2 – Essentially radiative models coupled with other models

These are physical models where a part of the radiative flux is absorbed inside the transparent or semi-transparent propellant and becomes an energetic source. These models have to be coupled with other physical models to reach the combustion rate. At first examination, it could appear that at least three models could possibly be used to explain, for example a doubled combustion rate under ETC i.e.:

a – a bulk heating model of the propellant as mentioned by Wren and al. [8] coupled with a classical ablative model as mentioned before, where the bulk heating increases the initial temperature of ΔTo, to get To^* the initial temperature for the action of the remaining energetic flux. For doubling of the combustion rate, if the energetic flux remains constant, the ablation energy is just divided by 2, so the bulk heating is $\Delta To = 208.5$ K or the initial temperature of the propellant is $To^* = 501.5$ K.

b – a bolk heating model of the surface layer of the propellant whose supposed Arrhenius type decomposition reaction imposes its kinetic rate to the combustion, as for low pressure combustion or as mentioned by Lowe [4]. The surface temperature for classic combustion Ts will increase to Ts^* by radiative bulk-heating $\Delta Ts = Ts^* - Ts$. Numerically for a doubled ETC combustion rate and for a temperature of say, $Ts = 710$ K for classical combustion, the warming-up of the surface layer ΔTs due to extra ETC radiative flux is a function of Arrhenius activation energy: 10 kJ/mol $\Rightarrow \Delta Ts = 147$ K, 100 kJ/mol $\Rightarrow \Delta Ts = 11$ K and 400 kJ/mol $\Rightarrow \Delta Ts = 3$ K.

c – a modification in the vicinity of the propellant surface by local radiative absorption which creates defects as suggested by several US and German works, and particularly by the results presented by Weise and al. [9] showing for an uncoated propellant an enhancement of roughly six times the combustion rate of that of a carbon coated propellant, when both are burning with ETC. A possible explanation is that if the propellant with its defects becomes equivalent to a porous propellant as presented by ICT [6], its real surface in ablation is increased. Furthermore if we assume that the intrinsic combustion rate is not modified by ETC combustion, the ratio of ETC to classical combustion rate is the same as the ratio of the actual surface modified by defects generated by radiative absorption to the classical combustion surface.

3 – Combustion behaviour models

We also applied the results and their analysis by Avi Birk and al. [10] in the vicinity of plasma injection, to our case. This experimental work suggests that a part of the propellant may react quicker than the rest of the charge. The pressure derivative curve versus pressure, for a loading density of 0.3 g/cm^3 for classic ignition and ETC ignition, suggests that the ETC ignition pressure derivative may be decomposed in two parts: 80% intact grains and the remaining 20% of plasma perturbed grains. The question then is to know what is the plasma perturbation of the grain. Further elements to estimate the perturbation may be given by the experimental combustion surface of the propellant given in Figure 4 for classic ignition (bold line) and ETC ignition (thin line). This figure shows that the ETC combustion surface may be decompose into two parts: one part of 80% of classically burning grain surface (black dots), and the remaining 20% of plasma perturbed grain (PPG) surface (triangles). As the figure shows, the latter surface – proportional to experimental form function of PPG – seems to be almost linearly decreasing to zero in the range 50 to 300 MPa.

4 – Semi-empirical relation for classic and ETC ignition

The ISL model for a LOVA propellant proposed by Grune and al. [11] applied to our case – even if our propellant is different – allows the calculation of the temperature TETC of the gas phase for ETC firings. This temperature compared with numerical simulation gives us another element which confirms the combustion of GP during the injection of plasma in this range of plasma energy.

The DERA model mentioned by Firth and al. [12] multiplies the classical combustion rate by a linear expression of electrical power. For our case we tried this expression as a first approximation during the ETC ignition phase only – which is the scope of our research. In our configuration where the plasma is not generated in situ but in the torch, the electrical power acting on the propellant under plasma form may be considered as a simple time delay and allows us to obtain a similar linear expression as that obtained by DERA.

CONCLUSION

The STANAG 4115 method that we adapted to ETC is usable to determine experimental combustion rate and therefore to easily characterise a propellant for industrial purposes by its combustion rate in the configuration tested. The extrapolation of these combustion rate results to real weapon systems with much higher loading densities has to be carefully carried out, as pointed out by Bashung and al. [13].

The use of numerous existing possible physical models allows several explanations for the same experimental combustion rate. These explanations in the presented case obtained with the above models are puzzling as valid for ratios of ETC to classic combustion rate ranging from 1 (unmodified) to almost double and over (enhanced).

This is understandable as the ETC ignition differs considerably from classical ignition where the temperature and the composition of the gas phase remains almost constant during the firing. The first numerical simulation with coupling of thermodynamic equilibrium calculation shows (1) the large variation of temperature and composition of the gas phase in ETC condition (2) the inadequacy of the STANAG method to obtain the thermal losses to the wall.

All these points shows the necessity to clarify the phenomena involved by further experimentation and analysis.

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